Specific heat of MgB_2 after irradiation

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Abstract. We studied the effect of disorder on the superconducting properties of polycrystalline MgB_2 by specific-heat measurements. In the pristine state, these measurements give a bulk confirmation of the presence of two superconducting gaps with $2\Delta_0/k_B T_c = 1.3$ and 3.9 with nearly equal weights. The scattering introduced by irradiation suppresses T_c and tends to average the two gaps although less than predicted by theory. We also found that by a suitable irradiation process by fast neutrons, a substantial bulk increase of dH_{c2}/dT at T_c can be obtained without sacrificing more than a few degrees in T_c. The upper critical field of the sample after irradiation exceeds 28 T at T→ 0.

1. Introduction

The recently discovered superconductor MgB$_2$ has attracted a lot of attention, because of its unexpectedly high critical temperature, T_c, for a phonon-mediated pairing mechanism [1, 2, 3, 4, 5, 6]. It has been proposed that such a high T_c is due to the existence of two superconducting gaps [2, 3], with this claim supported by experiments [5, 6]. It was suggested that the two gaps open on different parts of the Fermi surface [2, 3]. One part is three-dimensional, and arises from the π bonding and antibonding orbitals; it gives rise to the smaller gap with $2\Delta_{\pi 0}/k_B T_c \sim 1.1 - 1.5$. The second part consists of nearly cylindrical sheets, which arise from the σ-band, and gives rise to the larger gap with $2\Delta_{\sigma 0}/k_B T_c \sim 3.6 - 4.5$.

The anisotropy of the superconducting gaps and that of the Fermi surface should lead to an anisotropic upper critical field, H_{c2}, and this has indeed been reported in MgB$_2$ [7, 8]. The anisotropy is temperature dependent, the ratio of H_{c2} in the boron plane to that along the c-axis being about 3 near T_c and about 7 at low temperature.

An empirical two-band model based on the presence of two different gaps was successfully used to fit specific-heat data obtained on different MgB$_2$ samples in zero field [9]. In this model, each gap is assumed to follow the usual temperature dependence of the BCS theory, whereas the gap ratio $2\Delta_0/k_B T_c$ is allowed to differ from the BCS value 3.5. Both gaps are expected to be sensitive to impurity scattering [2]. An interesting prediction is that non-magnetic interband scattering will decrease T_c in the case of two coupled gaps, whereas Anderson's theorem would rather predict that scattering is irrelevant to first order for a single gap [10]. In the limit of strong scattering, it has been predicted that both gaps will be averaged up to the point where the single gap BCS limit is recovered. This should occur for $T_c \cong 27$ K [2]. This motivated the present study where the behavior of both gaps is followed by bulk specific-heat experiments while disorder is introduced in the material. We report heat capacity measurements on a polycrystalline sample before and after irradiation by fast neutrons. We found some suppression of the larger gap after irradiation, whereas the smaller gap remains quite robust.

Finally, the normal-state resistivity and the irreversibility field of MgB$_2$ were reported to be enhanced by proton irradiation [11]. It has also been shown that neutron irradiation can increase the upper critical field of this material [12]. Here we show that the upper critical field of our sample increases from about 18 to 28 T after irradiation. Compared to the results obtained on thin films by oxygen post-annealing [13] or fast quenching [14] techniques, the specificity of this study lies in the use of a bulk determination, in addition to magnetotransport measurements performed up to 28 T in a Bitter-type magnet.

2. Experimental details

The polycrystalline sample (HP14) was synthesized from Mg (99.8%) and B (99.7%) powders at 900°C for 1 hour in a cubic press at 3 GPa. Fig. 1(a) shows the diamagnetic transition of the pristine sample as detected by *ac* susceptibility at 8 kHz and 0.1 G. The midpoint of the transition is $T_c = 37$ K. Neutron irradiation was subsequently performed at the Triga research reactor in Vienna. The sample was irradiated to a fast neutron fluence (E>0.1 MeV) of 10^{22} m^{-2}, and in a second irradiation step a fluence of 2×10^{22} m^{-2} was added. Neutrons induce defects in MgB$_2$ mainly by neutron capture of ^{10}B followed by the emission of an alpha particle [12]. Due to the very small penetration range of low energy neutrons in MgB$_2$, thermal neutrons would induce damage only at the surface of the sample, resulting in a highly inhomogeneous defect structure. In order to avoid this problem, a cadmium shield was used, which absorbs nearly all low energy neutrons. The introduced disorder can be estimated to 1.7×10^{-2} and 5×10^{-2} dpa (displacements per atom) after the first and the second irradiation, respectively.

The transition temperature was measured after each irradiation by *ac* susceptibility as shown in Fig. 1. After irradiation, T_c shifted to lower temperature, 34.6 K after the first and 30.2 K after the second irradiation, and the transition broadened, but remained reasonably sharp compared to the result of similar studies [11]. Therefore the disorder introduced by neutron irradiation is fairly homogeneous.

The specific heat was measured before and after each irradiation in the temperature range 2 – 50 K by two different techniques. A relaxation calorimeter was used from 2 to 16 K [6], and an adiabatic technique from 16 to 50 K [15]. The resistivity of the sample was measured by the four-probe method. Data at fields below 17 T were obtained in a superconducting magnet by using a *dc* current reversal technique, with a current of 2 mA. Additional measurements from 12 to 28 T were performed in a Bitter coil at the GHMFL in Grenoble using an *ac* synchronous detection technique. These measurements were performed with a current of 2 mA at 11.7 Hz.

3. Results and discussion

3.1. The increase of H_{c2}

The specific heat of the sample was measured before and after each irradiation in different magnetic fields. Fig. 2 shows the specific heat difference $[C(B) - C(14\mathrm{T})]/T$. The baseline $C(14\mathrm{T})$ represents the normal state specific heat in the temperature range shown in Fig. 2; it was smoothed before subtraction. Since the lattice contribution cancels after subtraction, Fig. 2 in fact shows the difference between the electronic specific heat in the superconducting and the normal state. The amplitude of the specific-heat jump in zero field decreases after each irradiation. Its position is shifted in bulk by the magnetic field to a lower temperature. The broadening of the transition in increasing fields results from angular averaging since our sample is polycrystalline and anisotropic. In MgB$_2$, the anisotropy of the H_{c2} is estimated as 2 – 3 close to T_c [6, 7, 8].

Because of the transition broadening due to anisotropy, the onset of the specific-heat anomaly (see arrows in Fig. 2) was used to define T_c. This would correspond to the transition temperature of a single crystal for H parallel to the boron planes. At $H = 0$, the critical temperature is 37.4 K, 35.8 K and 31.2 K for the sample before, after the first and after the second irradiation respectively. Fig. 3 shows the temperature dependence of H_{c2} obtained from both specific-heat and resistance measurements (before and after irradiation). Note that the determinations based on the specific-heat jump are no longer significant at high fields owing to the smearing of the jump. Nevertheless, they unambiguously demonstrate that the average initial slope $(dH_{c2}/dT)_{T_c}$ increases after each irradiation.

A positive curvature (PC) of H_{c2} near T_c can been seen in Fig. 3 at different stages of irradiation. This can be attributed to the existence of two superconducting gaps in the sample, which may be considered as a particular case of gap anisotropy [16, 17]. It is pointed out by theory [17, 19] that the PC disappears in the "dirty" limit, where very strong interband scattering suppresses the anisotropy. In order to investigate the change of PC after irradiation, we used the following formula to fit our data near T_c up to 4 - 5 T: $H_{c2}(T) = H_{c2}^{\star}(1 - T/T_c)^{\alpha}$ [18], where T_c is the transition temperature at zero field, $H_{c2}^{\star}$ and α are fitting parameter. We found α within the range of 1.4 ± 0.1 before and after each irradiation, and no significant trend was observed. This is consistent with the observation that both gaps can still be identified after irradiation, confirming that the interband scattering caused by disorder in this sample is not strong enough to reach the "dirty" limit, as discussed in Section 3.2.

In order to verify the increase of H_{c2} after irradiation, we performed resistance measurements in magnetic field up to 17 T on the pristine sample and up to 28 T after the second irradiation. The insets of Fig. 3 show typical field and temperature sweeps after the second irradiation. For the pristine sample, $H_{c2}(0)$ is estimated to be about 18 T, in agreement with the results obtained on single crystals for the field parallel to the boron plane [7, 29]. After the second irradiation, the transition becomes broader at low temperature, as it was already the case for the pristine sample, presumably again as a consequence of anisotropy [7]. At $T = 1.5$ K, the transition is not complete even at 28 T, indicating $H_{c2}(0)$ higher than 28 T. Both specific-heat onsets and resistance transition midpoints are plotted in the $H - T$ phase diagram of Fig. 3. The results of both determinations of $H_{c2}(T)$ agree reasonably well in the low field range, the onset of calorimetric transitions appearing as an upper limit as expected. The quasi-linear increase of $H_{c2}(T)$ down to $T = 0$ differs remarkably from the horizontal slope at $T \to 0$ found for conventional superconductors. An explanation for this unusual behavior was provided within the two band model [16]. Note that a similar PC was found in several borocabides [18], not necessarily implying the same physical origin [19].

For a type-II superconductor in the dirty limit, $H_{c2}(0) \propto \gamma_n \rho T_c$, where γ_n is the Sommerfeld coefficient in the normal state, and ρ is the normal-state resistivity at low temperature [20]. After irradiation, T_c decreases by 20%, so that the main parameters to be investigated for changes are γ_n and ρ. As will be shown later, γ_n does not change

significantly.

The residual resistivity depends sensitively on disorder. Before irradiation, just above T_c, $\rho(45\text{K}) = 4.0 \ \mu\Omega.\text{cm}$ and the residual resistance ratio is $RRR = \rho(300\text{K})/\rho(45\text{K}) = 3.3$. To estimate the electronic mean free path, we use the following formula:

$$l = [\frac{\pi k_\text{B}}{e}]^2 \cdot \frac{\sigma}{\gamma_n v_\text{F}}, \tag{1}$$

where σ is the normal-state conductivity and v_F is the Fermi velocity. With $\gamma_n = 0.14$ mJ/K^{2}cm^3 [21] and $v_\text{F} = 4.8 \times 10^7$ cm/s [22], this yields $l \approx 270$ Å. Since $l \gg \langle \xi \rangle \approx 42$ Å [23], the sample is initially rather in the clean limit. After the second irradiation, the low-temperature resistivity increases to $\rho(45\text{K}) = 22.6\mu\Omega.\text{cm}$ (RRR = 1.4), which gives $l \approx 50$ Å. The sample is now in the intermediate case between the clean and dirty limit. In the dirty limit, the upper critical field would be determined by the geometrical average between the coherence length and the mean free path $\xi = \sqrt{\xi_0 l}$. Therefore, at least qualitatively, one is led to conclude that disorder induced by irradiation affects H_{c2} through the reduction of the mean free path. However, we note that the sample approaches the dirty limit only after the irradiation, so that the discussion given here remains essentially qualitative. Moreover, the two-band nature of MgB$_2$ complicates the determination of the mean free path [26].

Fig. 4 shows the specific heat below 16 K in different magnetic fields at different stages of irradiation. At the highest field accessible to our measurements, $\mu_0 H = 14$ T, C/T reflects nearly the normal-state specific heat. At first view, this would not seem to be the case, since $\mu_0 H_{c2}(0)$ is 18 and 28 T before and after the second irradiation, respectively. However, it was shown by several experiments that γ versus H already saturates at about $H_{c2}/2$ in polycrystals [6, 24], so that no significant change is expected above 14 T. This is also illustrated by the behavior of the pristine sample (see Fig. 4(a)), where specific heat saturates near 8 T. In the inset of Fig. 4, we show that the low temperature specific heat at 14 T remains unchanged at different stages of the irradiation, indicating that the normal state Sommerfeld coefficient does not change (the downturn that occurs at very low temperature in the C/T versus T^2 plot in the inset of Fig. 4 is believed to be due to the presence of residual magnetic impurities). Therefore, as already mentioned above, the variation of H_{c2} is not due to a change in γ_n. We thus conclude that irradiation merely enhances the scattering, causing both a decrease of T_c and an increase of the upper critical field.

These results suggest that other methods able to introduce disorder, such as fast quenching or chemical doping, could also serve to increase H_{c2}, as was realized in A15 compounds [25]. Indeed, a large increase of H_{c2} was obtained by metallurgical heat treatment of MgB$_2$ thin films [13, 14]. The present study shows that this increase of H_{c2} is a bulk property, at least at low fields, where the specific-heat jump is clearly observable. It is confirmed by direct resistance measurements at low temperatures and high magnetic fields, rather than by extrapolation beyond 10 T in previous work.

3.2. Effect of irradiation on the gaps

As discussed above, irradiation does not reduce the density of states, but merely increases the scattering. For a one-band, isotropic superconductor, scattering due to non-magnetic impurities would account for the increase of the normal-state resistivity, but would not affect T_c to first order. For a two-band superconductor, *intra*band scattering would preserve the two-gap structure even for a high concentration of impurities [26]. In the present case, a clear suppression of T_c is observed; it can only be explained by *inter*band scattering between the σ- and π- bands [2, 10]. Therefore, a distinction should be made between samples for which impurities affect only the normal-state resistivity, leaving T_c unchanged [26], and the present irradiated samples, for which both T_c and resistivity are affected. These distinct situations reflect different balances between intra- and interband scattering. If we believe that strong interband scattering exists for the present samples, which is rather unusual for MgB$_2$ [26], we expect changes in the two-gap structure after strong irradiation. We proceed to study this point experimentally.

The electronic specific heat C_e/T versus T at zero field is plotted in Fig. 5. The lattice contribution determined at 14 T was smoothed and subtracted. The low temperature excess of the specific heat with respect to a one-gap BCS curve (hatched area in Fig. 5), which results from the existence of the smaller gap [9], becomes less pronounced after irradiation. A satisfactory fit can be obtained for the unirradiated sample using a two-gap model [9, 21]. The fit gives $\Delta_0 = 6.1$ meV for the larger gap and 2.1 meV for the smaller gap, with nearly equal weights. Similar fits can be made after irradiation. The gap values at each stage of irradiation are given in Table 1. After the second irradiation, the quality of the fit was degraded by the broadening of the transition. A satisfactory fit was obtained by shifting T_c slightly above the midpoint of the jump and by lowering γ_n by 2.5%.

An obvious effect of the irradiation is the suppression of the larger gap, which, in a two-gap model, is reflected in the specific heat jump at T_c [9]. This gap, which originates from the 2D σ-band, seems to be more sensitive to defects. The smaller gap, which is reflected in the low temperature hump in the specific heat near 10 K, appears to be quite robust in absolute value, remaining within 10% of the average 2.15 meV in all cases. However, its reduced value $2\Delta_0/k_B T_c$ increases after each irradiation, due to the decrease of T_c. Liu et al. predicted that interband scattering in MgB$_2$ should average both gaps, which would finally merge into a single BCS gap when T_c reaches ~ 27 K [2]. The observed gap values are plotted as a function of T_c in the inset of Fig. 5. This plot suggests that the single-gap limit will only be reached for T_c smaller than the anticipated 27 K, although additional irradiations will be required to prove it. From the fits, one can also see that the weight of both gaps remains almost unchanged after the irradiation.

Golubov et al. calculated the effect of interband scattering on specific heat due to non-magnetic impurities within the two-band model [10], and pointed out that

the impurity scattering should increase the $\Delta C/\gamma_n T_c$ ratio, whereas the specific heat jump ΔC should remain nearly constant. According to our experiment, ΔC decreases after irradiation and $\Delta C/\gamma_n T_c$ decreases only slightly. This disagreement may be of experimental origin and due to the transition broadening.

The coefficient of the mixed-state electronic specific-heat, $\gamma(H)$, provides independent information on both gaps. Its highly non-linear increase is shown by the plot $C(H,T)/T$ at $T \ll T_c$ (here, 3K) on a logarithmic field scale (see Fig. 6). The curves at different stages of irradiation lie nearly parallel to each other below 10 T, and saturate at the same value of γ_n at 14 T. The small maximum at 8 T for the non-irradiated sample is believed to be an artifact due to a residual Schottky contribution of magnetic origin. The fact that the three curves are parallel to each other implies a simple relation $\gamma_{\text{pristine}}(H) = \gamma_{\text{irradiated}}(\alpha H)$, where $\alpha \approx 1.7$ after the first irradiation and $\alpha \approx 2$ after the second one. The inset of Fig. 6 illustrates this relation: the C/T versus T^2 curves at 0.5 T for the pristine sample, 0.84 T after the first irradiation, and 1 T after the second irradiation point to the same value of $\gamma(H, T = 0)$.

According to theory [2] and experiment [9, 27], both gaps have nearly equal weights, i.e. the partial densities of states N_σ and N_π of the associated bands, and the partial normal-state Sommerfeld coefficients $\gamma_{n\sigma}$ and $\gamma_{n\pi}$, are nearly equal. Recent scanning tunneling microscopy measurements on a single crystal show that already in a magnetic field of 0.2 T, there is a significant overlap of vortex cores in the π-band vortices [28]. Very recent specific-heat measurements on a single crystal have revealed that $\gamma_\pi(H)$ increases very fast with the field, and saturates above a crossover field $\mu_0 H \cong 0.4$T, whereas $\gamma_\sigma(H)$ saturates at 3.3 T (field along the c-axis) or 18 T (field in the ab-plane) [29]. It follows that at low fields($B \leq 0.5$ T) the contribution from the smaller gap dominates the behavior of $\gamma(H)$, in agreement with theory [30]. On the contrary, at high fields ($B > 1$ T), the contribution from the smaller gap saturates, so that the variation of $\gamma(H)$ comes from the larger gap. The crossover near 0.4 T can be considered as the virtual upper critical field $H_{c2\pi}$ for the smaller gap. The parallelism of the curves in Fig. 6 at high fields ($1 - 8$ T) provides the proof for the increase of H_{c2} associated with the larger gap. This is independently measured by the shift of the specific-heat jump and resistive transitions. Alternatively, at low fields ($B < 0.5$ T), below the saturation of the contribution $\gamma_\pi(H)$ associated with the smaller gap, one may deduce from the approximate relationship $\gamma_\pi(H) = \gamma_{n\pi} H/H_{c2}$ that $H_{c2\pi}$ has increased after irradiation by the same factor α as $H_{c2\sigma}$. This observation tends to support the interpretation that the superconductivity in the π-band is not independent, but rather induced by superconductivity in the σ-band [30, 28]. At this point we must recall that our sample is a polycrystal, so that information on the anisotropy is lost in the high field region dominated by the large gap of the σ-band. More detailed information on this unusual system would require irradiation experiments on single crystals.

4. Conclusion

We studied irradiation effects on a polycrystalline sample of MgB$_2$. Irradiation by fast neutrons provides a good tool to introduce *inter*band scattering. Specific-heat measurements show a suppression of the larger gap after irradiation, whereas the smaller gap Δ_π remains nearly unchanged. In terms of reduced gap value $2\Delta_0/k_B T_c$, the gaps tend to converge, but slower than predicted by considerations of the interband scattering. The two-gap feature remains quite robust. We also find that the upper critical or crossover field $H_{c2\pi}$ associated with the smaller gap follows the behavior of that of the larger gap after irradiation. Together with resistivity measurements, we show that a substantial increase of H_{c2} can be obtained by irradiation, without sacrificing more than a few degrees in T_c. If critical currents do not collapse, the potential of MgB$_2$ tuned by disorder is worth further investigations in view of high field superconducting applications.

Acknowledgments

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	Before irradiation	After 1st irradiation	After 2nd irradiation
T_c (K)	37	34.1	30.2
γ_n (mJ/K^2.gat)	0.83	0.83	0.81
Δ_π(meV)	2.07 ± 0.20	2.20 ± 0.20	2.22 ± 0.20
$2\Delta_\pi/k_B T_c$	1.3	1.5	1.7
Δ_σ(meV)	6.21 ± 0.30	5.30 ± 0.25	4.68 ± 0.35
$2\Delta_\sigma/k_B T_c$	3.9	3.6	3.6
$\gamma_{n\pi} : \gamma_{n\sigma}$	0.5:0.5	0.55:0.45	0.55:0.45
ΔC(mJ/K.gat)	27.6 ± 1	25.3 ± 1	20.3 ± 1
$\Delta C/\gamma_n T_c$	0.9	0.85	0.85

Table 1. Gap parameters of MgB$_2$ at different stages of irradiation obtained by fits of the specific heat. γ_n is the normal-state Sommerfeld coefficient. Δ_π and Δ_σ are the gap values for the smaller and larger gap. ΔC is the specific-heat jump. $\gamma_{n\pi}$ and $\gamma_{n\sigma}$ are the partial Sommerfeld coefficients associated with the π- and σ-band, respectively; $\gamma_{n\pi} + \gamma_{n\sigma} = \gamma_n$.

Figure captions

Figure 1. Diamagnetic transition of the sample measured by *ac* susceptibility, real (χ') and imaginary (χ'') part: (a) before irradiation; (b) after the first irradiation; (c) after the second irradiation.

Figure 2. Transitions at T_c in different magnetic fields measured by specific heat, $\Delta C = C(B = 0) - C(B = 14\text{T})$: (a) before irradiation, from right to left $B = 0$, 0.5, 1, 2, 3 T, (b) after the first irradiation, from right to left $B =$ 0, 0.84, 1.7, 2.5, 3.4, 4.2, 5.1 T, (c) after the second irradiation, from right to left $B =$ 0, 0.5, 1, 2, 3, 4, 5, 6, 8 T. Note that the sample was cut and its mass reduced for the irradiations, explaining the increased scatter.

Figure 3. Upper critical field $H_{c2}(T)$ before and after irradiation: $\triangle$ and full line, before the irradiation, determined by the onset of the specific-heat jump; $\square$, before the irradiation, determined by the midpoint of the resistance step, $\diamond$ and full line, after the first irradiation, specific heat onset; $\circ$ and full line, after the second irradiation, specific heat onset; $\bullet$, after the second irradiation, transition midpoint in resistance. Inset: resistance measurements after the second irradiation: upper right: magnetic field sweeps at constant temperature, from left to right: $T = 28$, 24, 20, 16, 12, 8, 4, 1.5 K; lower left: temperature sweeps at constant field, from left to right: $B = 14$, 12.5, 10, 8.5 T.

Figure 4. Total specific heat below 16 K in a C/T versus T^2 plot: (a) before irradiation, from bottom to top: $B =$ 0 , 0.05, 0.1, 0.2, 0.3, 0.5, 1, 2, 4, 8, and 14 T; (b) after the first irradiation, from bottom to top: $B =$ 0, 0.17, 0.5, 0.84, 1.7, 3.4, 6.8, 14 T; (c) after the second irradiation, from bottom to top: $B =$ 0, 0.2, 0.5, 0.75, 1, 2, 4, 8 T. Inset: low temperature specific heat at 14 T before and after each irradiation (three data sets). The extrapolation to T = 0 shows that γ_n does not vary.

Figure 5. Electronic specific heat C_e/T versus T with two-gap fits [9], from top to bottom: before irradiation, after the first irradiation, after the second irradiation, respectively. The latter two curves are shifted by 0.25 mJ/K^{2}gat for clarity. The dashed line represents the BCS single-gap model with the same T_c and γ_n as the sample after the second irradiation. Inset: variation of both gaps as a function of T_c (note that the scale is reversed to show increasing scattering from left to right). The dashed line is the isotropic BCS limit. The star $\star$ is a calculated point where the gaps should converge to the BCS value according to Ref. [2].

Figure 6. Specific heat C/T at 3 K versus magnetic field on a logarithmic scale, $\diamond$ before irradiation, $\bullet$ after the first irradiation, $\triangle$ after the second irradiation. Inset: from bottom to top, low temperature specific heat at 0.5 T before irradiation, 0.84 T after the first irradiation and 1 T after the second irradiation, respectively.
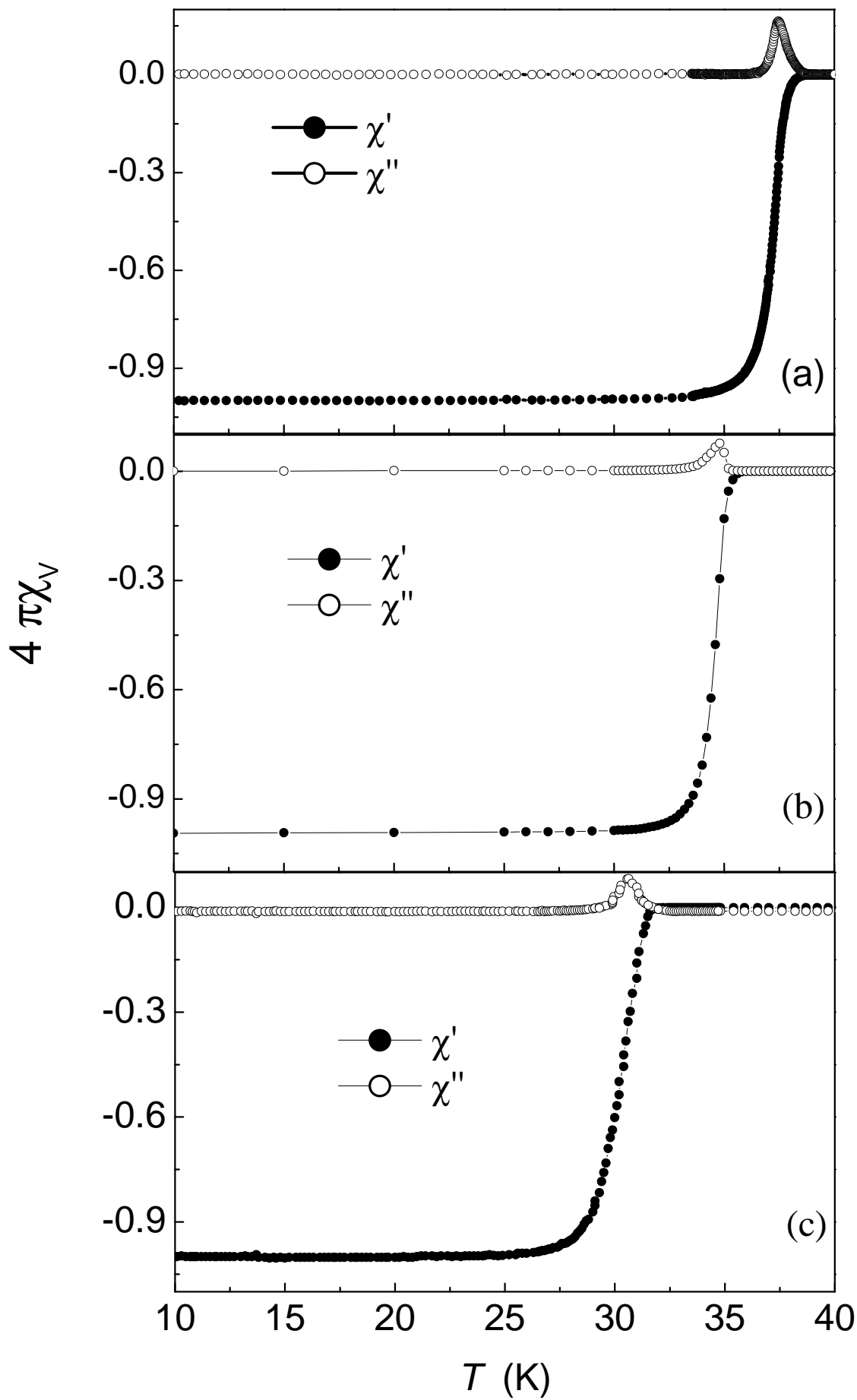
This figure "Fig2.JPG" is available in "JPG" format from:

http://arXiv.org/ps/cond-mat/0208169v3
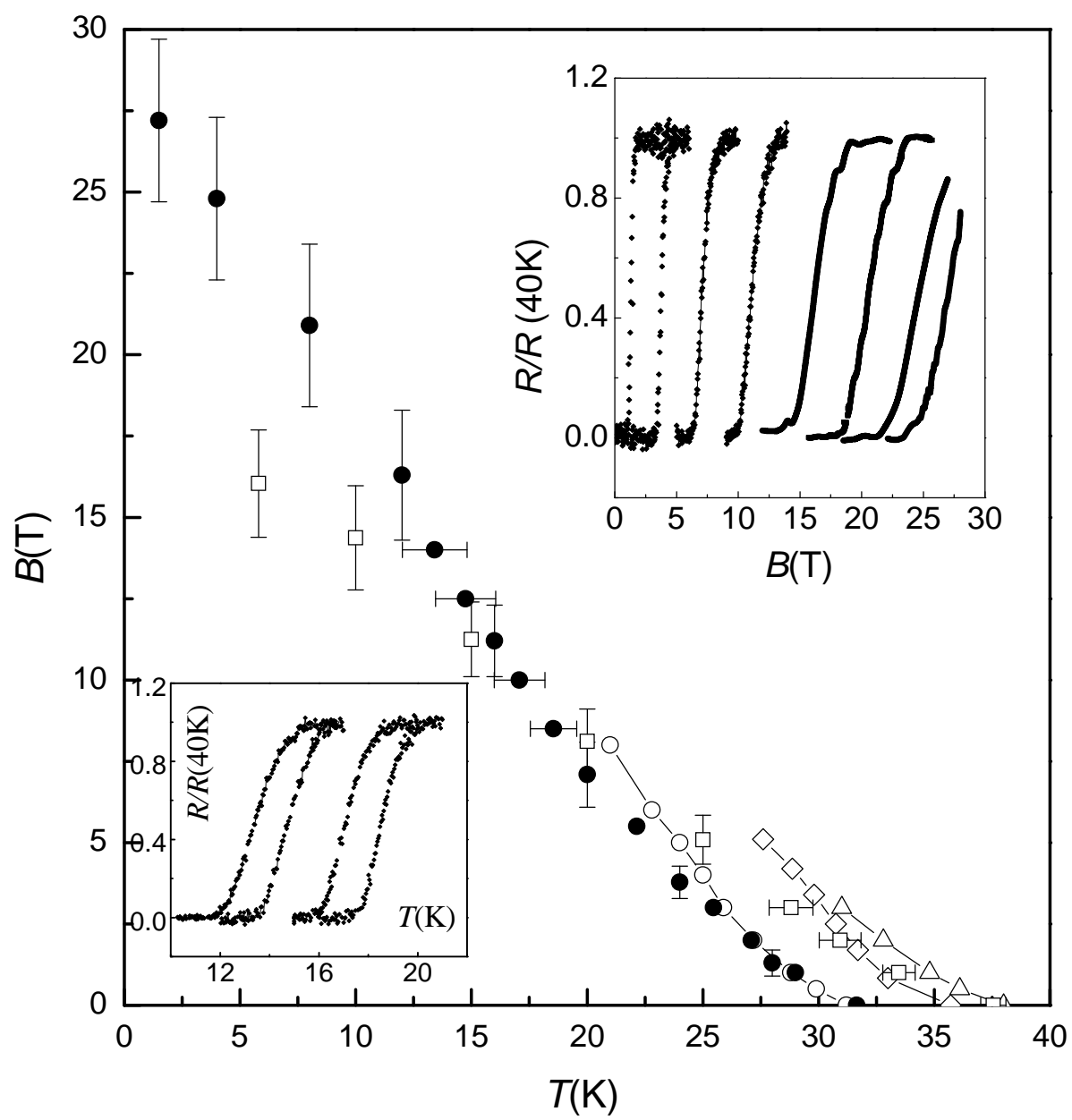
This figure "Fig4.JPG" is available in "JPG" format from:

http://arXiv.org/ps/cond-mat/0208169v3